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TRANSMITTAL LETTER TO THE UNITED STATES

DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER 49741

09/889404

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

INTERNATIONAL APPLICATION NO. INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED
PCT/EP 00/00911 5 February 2000 9 February 1999

TITLE OF INVENTION: FUEL COMPOSITION

APPLICANT(S) FOR DO/EO/US Harald SCHWAHN, Dietmar POSSELT

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. /X/ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
 2. // This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
 3. /X/ This express request to begin national examination procedures (35 U.S.C.371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
 4. /x/ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. /X/ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a./X/ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b.// has been transmitted by the International Bureau.
 - c.// is not required, as the application was filed in the United States Receiving Office (RO/USO).
 6. /X/ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 7. // Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).
 - a.// are transmitted herewith (required only if not transmitted by the International Bureau).
 - b.// have been transmitted by the International Bureau.
 - c.// have not been made; however, the time limit for making such amendments has NOT expired.
 - d.// have not been made and will not be made.
 8. // A translation of the amendments to the claims under PCT Article 19(35 U.S.C. 371(c)(3)).
 9. /x/ An oath or declaration of the inventor(s)(35 U.S.C. 171(c)(4)).
 10. // A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:
11. / / An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. /x/ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. /x/ A FIRST preliminary amendment.
// A SECOND or SUBSEQUENT preliminary amendment.
 14. // A substitute specification.
 15. // A change of power of attorney and/or address letter.
 16. /x/ Other items or information.
International Search Report
International Preliminary Examination Report

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49741

	<u>CALCULATIONS</u>	<u>PTO USE ONLY</u>
17. /X/ The following fees are submitted BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)); Search Report has been prepared by the EPO or JPO.....\$860.00	860.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482).....\$750.00		
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....\$700.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO\$ 970.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied pro visions of PCT Article 33(2)-(4).....,\$96.00		

ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration
later than // 20 // 30 months from the earliest
claimed priority date (37 CFR 1.492(e)).

Claims	Number Filed	Number Extra	Rate	
Total Claims	14	-20	X\$18.	
Indep. Claims	1	-3	X\$80.	
Multiple dependent claim(s)(if applicable)		+270.		
TOTAL OF ABOVE CALCULATION		=	860.00	
Reduction of 1/2 for filing by small entity, if applicable.				
Verified Small Entity statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).				
		SUBTOTAL	=	860.00
Processing fee of \$130. for furnishing the English translation later than // 20 // 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				
		+		
		TOTAL NATIONAL FEE	=	860.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) \$40.00 per property				
		=	40.00	
		TOTAL FEES ENCLOSED	=	\$ 900.00

Amount to be
refunded: \$ _____
Charged \$ _____

- a./X/ A check in the amount of \$ 900. to cover the above fees is enclosed.
- b.// Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c./X/ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 11-0345. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be filed and granted to restore the application to pending status.



SIGNATURE

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of)
SCHWAHN et al.) BOX PCT
)
International Application)
PCT/EP 00/00911)
)
Filed: February 5, 2000)
)

For: FUEL COMPOSITION

PRELIMINARY AMENDMENT

Honorable Commissioner of
Patents and Trademarks
Washington, D.C. 20231

Sir:

Prior to examination, kindly amend the above-identified application as follows:

IN THE CLAIMS

Kindly amend the claims as shown on the attached sheets.

R E M A R K S

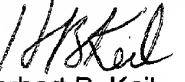
The claims have been amended to eliminate multiple dependency and to put the claim in better form for U.S. filing. No new matter is included.

A clean copy of the claims is attached.

Favorable action is solicited.

Respectfully submitted,

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CLEAN VERSION OF AMENDED CLAIMS - OZ 49741

11. A fuel composition as defined in claim 1, containing a gasoline having an olefin content of not more than 21 vol%.
12. A fuel composition as defined in claim 1, containing a gasoline having a benzene content of not more than 1.0 vol%.
13. A fuel composition as defined in claim 1, containing a gasoline having an oxygen content of not more than 2.7 wt%.
14. A fuel composition as defined in claim 1, containing the gasoline additives containing the polar groups (a) to (i) in a concentration of from 1 to 5000 ppm by weight.

MARKED UP VERSION OF AMENDED CLAIMS - OZ 49741

11. A fuel composition as defined in claim 1 [any of claims 1 to 10], containing a gasoline having an olefin content of not more than 21 vol%.
12. A fuel composition as defined in claim 1 [any of claims 1 to 11], containing a gasoline having a benzene content of not more than 1.0 vol%.
13. A fuel composition as defined in claim 1 [any of claims 1 to 12], containing a gasoline having an oxygen content of not more than 2.7 wt%.
14. A fuel composition as defined in claim 1 [any of claims 1 to 13], containing the gasoline additives containing the polar groups (a) to (i) in a concentration of from 1 to 5000 ppm by weight.

CURRENT CLAIMS - OZ 49741

1. A fuel composition containing, as major component, a gasoline having an aromatics content of not more than 42 vol% and a sulfur content of not more than 150 ppm by weight, and, as minor component, at least one gasoline additive having a detergent action or an anti-valve-seat-wear action, wherein this gasoline additive contains at least one hydrophobic hydrocarbon group having a number-average molecular weight (M_n) of from 85 to 20,000 and at least one polar group selected from
- (a) monoamino or polyamino groups containing up to 6 nitrogen atoms, of which at least one has alkaline properties,
 - (b) nitro groups, optionally combined with hydroxyl groups,
 - (c) hydroxyl groups combined with monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties,
 - (d) carboxylic acid groups or the alkali metal or alkaline earth metal salts thereof,
 - (e) sulfo groups or the alkali metal or alkaline earth metal salts thereof,
 - (f) polyoxy-(C_2-C_4 alkylene) groups which are terminated by hydroxyl groups, monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties, or by carbamate groups,
 - (g) carboxylate groups,
 - (h) groups derived from succinic anhydride and containing hydroxyl and/or amino and/or amido and/or imido groups and
 - (i) groups produced by Mannich reaction of substituted phenols with

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aldehydes and mono- or poly-amines.

2. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (a), polyalkene monoamine or polyalkene polyamines based on polypropylene, polybutylene or polyisobutylene having a molecular weight M_n of from 300 to 5000.
3. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (b), reaction products of polyisobutenes having an average degree of polymerization P of from 5 to 100 with nitrogen oxides or mixtures of nitrogen oxides and oxygen.
4. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (c), reaction products of polyisobutene epoxides, obtained from polyisobutylene containing predominantly terminal double bonds apd having a molecular weight M_n of from 300 to 5000, with ammonia, mono- or poly-amines.
5. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (d), copolymers of C_2-C_{40} olefins with maleic anhydride having a total molecular weight of from 500 to 20,000 whose carboxylic acid groups are completely or partially converted to the alkali metal or alkaline earth metal salts and the remainder of the carboxylic acid groups has been caused to react with an alcohol or amine.
6. A fuel composition as defined in claim 1, containing, as gasoline additive

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containing polar groups (e), an alkali metal or alkaline earth metal salt of an alkyl sulfosuccinate.

7. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (f), a polyether or polyether amine, obtainable by reaction of a C₂-C₃₀ alkanol, C₆-C₆₀ alkanediol, mono- or di-(C₂-C₃₀ alkyl)amine, C₁-C₃₀ alkylcyclohexanol or C₁-C₃₀ alkylphenol with from 1 to 30 mol of ethylene oxide and/or propylene oxide and/or butylene oxide per hydroxyl group or amino group and, in the case of polyether amines, by subsequent reductive amination with ammonia, a monoamine or a polyamine.
8. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (g), a, ester of a mono-, di- or tri-carboxylic acid with a long-chain alkanol or polyol.
9. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (h), a derivative of polyisobutensylsuccinic anhydride, obtained by reaction of conventional or highly reactive polyisobutylene having a molecular weight M_n of from 300 to 5000 with maleic anhydride by thermal treatment or via the chlorinated polyisobutylene.
10. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (i), a reaction product of a polyisobutene-substituted phenol with formaldehyde and a mono- or poly-amine.

CURRENT CLAIMS - OZ 49741

11. A fuel composition as defined in claim 1, containing a gasoline having an olefin content of not more than 21 vol%.
12. A fuel composition as defined in claim 1, containing a gasoline having a benzene content of not more than 1.0 vol%.
13. A fuel composition as defined in claim 1, containing a gasoline having an oxygen content of not more than 2.7 wt%.
14. A fuel composition as defined in claim 1, containing the gasoline additives containing the polar groups (a) to (i) in a concentration of from 1 to 5000 ppm by weight.

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Fuel Composition

- The present invention relates to a fuel composition containing,
5 as major component, a specific gasoline and, as minor component,
selected gasoline additives.

Carburetors and inlet systems of Otto engines, and also injection systems for fuel proportioning, are subjected to increasing
10 load due to contamination caused by dust particles from the air, unburned hydrocarbon residues from the combustion chamber and crankcase breather gases passed to the carburettor.

These residues shift the air-to-fuel ratio during idling and in
15 the lower partial load region, so that the mixture becomes leaner and combustion less complete and consequently the content of unburned or partly burned hydrocarbons in the exhaust gas increases and the gasoline consumption rises.

20 It is known to avoid these drawbacks by using fuel additives for cleaning the valves and carburetors or injection systems of Otto engines (cf eg: M. Rossenbeck in "Katalysatoren, Tenside, Miner-alöladditive", edited by J. Falbe, U. Hasserodt, page 223, G. Thieme Verlag, Stuttgart 1978).

25 Furthermore, the problem of valve seat wear occurs in the case of Otto engines of less recent design when fuelled with unleaded
gasolines. To counteract this, anti-valve-seat-wear additives based on alkali metal or alkaline earth metal compounds have been
30 developed.

For trouble-free running, modern Otto engines require automotive fuels having a complex set of properties which can only be guaranteed when use is made of appropriate gasoline additives. Such
35 gasolines usually consist of a complex mixture of chemical compounds and are characterized by physical parameters. The interrelationship between gasolines and appropriate additives in known fuel compositions is still unsatisfactory as regards their detergent action or their pollution-abating properties and their anti-
40 valve-seat-wear action.

It is thus an object of the present invention to provide a more effective gasoline/additive formulation.

45 Accordingly, we have found a fuel composition which contains, as major component, a gasoline having an aromatics content of not more than 42 vol% and a sulfur content of not more than 150 ppm

by weight, and, as minor component, at least one gasoline additive having a detergent action or an anti-valve-seat-wear action, which gasoline additive contains at least one hydrophobic hydrocarbon group having a number-average molecular weight (M_n) of from 5 85 to 20,000 and at least one polar group selected from

- (a) monoamino or polyamino groups containing up to 6 nitrogen atoms, of which at least one has alkaline properties,
 - 10 (b) nitro groups, optionally combined with hydroxyl groups,
 - (c) hydroxyl groups combined with monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties,
 - 15 (d) carboxylic acid groups or the alkali metal or alkaline earth metal salts thereof,
 - (e) sulfo groups or the alkali metal or alkaline earth metal salts thereof,
 - 20 (f) polyoxy-(C₂-C₄ alkylene) groups which are terminated by hydroxyl groups, by monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties, or by carbamate groups,
 - 25 (g) carboxylate groups,
 - (h) groups derived from succinic anhydride and containing hydroxyl and/or amino and/or amido and/or imido groups and
 - 30 (i) groups produced by Mannich reaction of substituted phenols with aldehydes and mono- or poly-amines.
- 35 The aromatics content of the gasoline is preferably not more than 40 vol% and more preferably not more than 38 vol%. Preferred ranges for the aromatics content are from 20 to 42 vol% and particularly from 25 to 40 vol%.
- 40 The sulfur content of the gasoline is preferably not more than 100 ppm by weight and more preferably not more than 50 ppm by weight. Preferred ranges for the sulfur content are from 0.5 to 150 ppm by weight and particularly from 1 to 100 ppm by weight.

In a preferred embodiment, the gasoline has an olefin content of not more than 21 vol%, preferably not more than 18 vol% and more preferably not more than 10 vol%. Preferred ranges for the olefin content are from 6 to 21 vol% and particularly from 7 to 18 vol%.

5

In another preferred embodiment, the gasoline has a benzene content of not more than 1.0 vol% and preferably not more than 0.9 vol%. Preferred ranges for the benzene content are from 0.5 to 1.0 vol% and preferably from 0.6 to 0.9 vol%.

10

In another preferred embodiment, the gasoline has an oxygen content of not more than 2.7 wt%, preferably from 0.1 to 2.7 wt%, more preferably from 1.0 to 2.7 wt% and most preferably from 1.2 to 2.0 wt%.

15

Particular preference is given to a gasoline which has an aromatics content of not more than 38 vol% and at the same time an olefin content of not more than 21 vol%, a sulfur content of not more than 50 ppm by weight, a benzene content of not more than 1.0 vol% and an oxygen content of from 1.0 to 2.7 wt%.

20

The content of alcohols and ethers in the gasoline is normally relatively low. Typical maximum contents are methanol 3 vol%, ethanol 5 vol%, isopropanol 10 vol%, tert-butanol 7 vol%, isobutanol 10 vol% and ethers containing 5 or more carbon atoms in the molecule 15 vol%.

The summer vapor pressure of the gasoline is usually not more than 70 kPa and preferably not more than 60 kPa (at 37°C).

30

The research octane number ("RON") of the gasoline is usually from 90 to 100. A usual range for the corresponding motor octane number ("MON") is from 80 to 90.

35

The above characteristics are determined by conventional methods (DIN EN 228).

The hydrophobic hydrocarbon group in the gasoline additives, which provides sufficient solubility in the fuel, has an average molecular weight (M_n) of from 85 to 20,000, preferably from 113 to 10,000 and more preferably from 300 to 5000. Typical hydrophobic hydrocarbon groups, particularly in conjunction with the polar groups (a), (c), (h) and (i), are polypropenyl, polybutenyl and polyisobutenyl radicals having molecular weights M_n of from 300 to 5000, preferably from 500 to 2500 and more preferably from 750 to 2250.

The following examples of individual gasoline additives having a detergent action or an anti-valve-seat-wear effect are mentioned by way of example.

- 5 Additives containing monoamino or polyamino groups (a) are preferably polyalkene monoamines or polyalkene polyamines based on polypropylene or highly reactive (ie containing predominantly terminal double bonds - mostly in the α and β positions) or conventional (ie containing predominantly centered double bonds) poly-
10 butylene or polyisobutylene having a molecular weight M_n of from 300 to 5000. Such additives based on highly reactive polyisobutylene which can be prepared from the polyisobutylene containing up to 20 wt% of *n*-butylene units, by hydroformylation and reductive amination with ammonia, monoamines or polyamines such as dimethyl-
15 laminopropylamine, ethylenediamine, diethylenetriamine, triethylenetetramine or tetraethylenepentamine, are disclosed, in particular, in EP-A 244,616. If the synthesis of the additives is based on polybutylene or polyisobutylene having predominantly centered double bonds (mostly in the β and γ positions) as starting materi-
20 als, an obvious choice is the synthesis method involving chlorination and subsequent amination, or oxidation of the double bond with air or ozone to form the carbonyl or carboxyl compound, with subsequent amination under reductive (hydrogenating) conditions. This amination may be carried out using the same amines as
25 mentioned above for the reductive amination of hydroformylated, highly reactive polyisobutylene. Corresponding additives based on polypropylene are described, in particular, in WO-A 94/24231.

Further preferred additives containing monoamino groups (a) are
30 the hydrogenation products of the reaction products of polyisobutylenes having an average degree of polymerization P of from 5 to 100 with nitrogen oxides or mixtures of nitrogen oxides and oxygen, as described, in particular, in WO-A 97/03946.

- 35 Further preferred additives containing monoamino groups (a) are the compounds produced from polyisobutylene epoxides by reaction with amines followed by dehydration and reduction of the amino alcohols, as described, in particular, in DE-A 196 20 262.
- 40 Additives containing nitro groups, optionally combined with hydroxyl groups (b), are preferably reaction products of polyisobutylenes having an average degree of polymerization P of from 5 to 100 or from 10 to 100 with nitrogen oxides or mixtures of nitrogen oxides and oxygen, as described, in particular, in WO-A
45 96/03367 and WO-A 96/03479. These reaction products are usually mixtures of pure nitropolyisobutanes (eg α,β -dinitropolyisobu-

tane) and mixed hydroxynitropolyisobutanes (eg α -nitro- β -hydroxy-polyisobutane).

Additives containing hydroxyl groups combined with monoamino or polyamino groups (c) are in particular reaction products of polyisobutylene epoxides, obtainable from polyisobutylene preferably containing predominantly terminal double bonds and having a molecular weight M_n of from 300 to 5000, with ammonia or mono- or poly-amines, as described, in particular, in EP-A 476,485.

Additives containing carboxylic acid groups or the alkali metal or alkaline earth metal salts thereof (d) are preferably copolymers of C_2-C_{40} olefins with maleic anhydride having a total molecular weight of from 500 to 20,000 whose carboxylic acid groups have been converted entirely or partially to the alkali metal or alkaline earth metal salts and the remainder of the carboxylic acid groups has been caused to react with alcohols or amines. Such additives are disclosed, in particular, in EP-A 307,815. Said additives mainly serve to prevent valve seat wear and can be used, as described in WO-A 87/01126, with advantage combined with conventional fuel detergents such as poly(iso)butylene amines or polyether amines.

Additives containing sulfo groups or the alkali metal or alkaline earth metal salts thereof (e) are preferably alkali metal or alkaline earth metal salts of an alkyl sulfosuccinate, as described, in particular, in EP-A 639,632. Such additives mainly serve to prevent valve seat wear and can be used with advantage combined with conventional fuel detergents such as poly(iso)butylene amines or polyether amines.

Additives containing polyoxy-(C_2-C_4 alkylene) groups (f) are preferably polyethers or polyether amines, which are obtained by reaction of C_2-C_{60} alkanols, C_6-C_{30} alkanediols, mono- or di-(C_2-C_{30} alkyl)amines, (C_1-C_{30} alkyl)cyclohexanols or (C_1-C_{30} alkyl)phenols with from 1 to 30 mol of ethylene oxide and/or propylene oxide and/or butylene oxide per hydroxyl group or amino group and, in the case of polyether amines, by subsequent reductive amination with ammonia, monoamines or polyamines. Such products are described, in particular, in EP-A 310,875, EP-A 356,725, EP-A 700,985 and US-A 4,877,416. In the case of polyethers such products also have flotation oil characteristics. Typical examples thereof are tridecanol butoxylates or isotridecanol butoxylates, isononylphenol butoxylates, polyisobutenol butoxylates and polyisobutenol propoxylates and the corresponding reaction products with ammonia.

Additives containing carboxylate groups (g) are preferably esters of mono-, di- or tri-carboxylic acids with long-chain alkanols or polyols, in particular those having a minimum viscosity of 2 mm²/s at 100°C, as described, in particular, in DE-A 3,838,918. The

- 5 mono-, di- or tri-carboxylic acids used can be aliphatic or aromatic acids, and suitable ester alcohols or ester polyols are primarily long-chain representatives containing, for example, from 6 to 24 carbon atoms. Typical representatives of these esters are adipates, phthalates, isophthalates, terephthalates and
10 trimellitates of isoctanol, isononanol, isodecanol and isotridecanol. Such products also have flotation oil characteristics.

Additives containing groups derived from succinic anhydride and containing hydroxyl and/or amino and/or amido and/or imido groups

- 15 (h) are preferably corresponding derivatives of polyisobutenyl succinic anhydride, which are obtained by reaction of conventional or highly reactive polyisobutylene having a molecular weight M_n of from 300 to 5000 with maleic anhydride by thermal treatment or via chlorinated polyisobutylene. Of special interest in this respect are derivatives with aliphatic polyamines such as ethylenediamine, diethylenetriamine, triethylenetetramine or tetraethylene-pentamine. Such gasoline additives are described, in particular, in US-A 4,849,572.

- 25 Additives containing groups (i) produced by Mannich reaction of substituted phenols with aldehydes and mono- or poly-amines are preferably reaction products of polyisobutylene-substituted phenols with formaldehyde and mono- or poly-amines such as ethylenediamine, diethylenetriamine, triethylenetetramine, tetraethylene-pentamine or dimethylaminopropylamine. The polyisobutenyl-substituted phenols can be derived from conventional or highly reactive polyisobutylene having a molecular weight M_n of from 300 to 5000. Such "polyisobutylene Mannich bases" are described, in particular, in EP-A 831,141.

- 35 To provide a more precise definition of the individual gasoline additives mentioned above, the disclosures of the aforementioned specifications of the prior art are included herein by reference.

- 40 The fuel composition of the invention can contain yet other conventional components and additives. Foremost examples thereof are flotation oils not having any marked detergent action, for example mineral flotation oils (base oils), in particular those of the viscosity class "Solvent Neutral (SN) 500 to 2000", and synthetic flotation oils based on olefin polymers having a molecular weight M_n of from 400 to 1800, mainly based on polybutylene or

polyisobutylene (hydrogenated or non-hydrogenated), on poly(α -olefin)s or poly(internal olefin)s.

Suitable solvents or diluents (for use in additive packs) are aliphatic and aromatic hydrocarbons, eg solvent naphtha.

Further conventional additives are corrosion inhibitors based, for example, on film-forming ammonium salts of organic carboxylic acids or heterocyclic aromatics for nonferrous metal corrosion

protection, antioxidants or stabilizing agents based, for example, on amines such as *p*-phenylenediamine, dicyclohexylamine or derivatives thereof or phenols such as 2,4-di-*tert*-butylphenol or 3,5-di-*tert*-butyl-4-hydroxyphenylpropionic acid, demulsifiers, antistatic agents, metallocenes such as ferrocene or methylcyclopentadienyl manganese tricarbonyl, lubricity additives such as specific fatty acids, alkenyl succinates, bis(hydroxylalkyl)fatty amines, hydroxyacetamide or castor oil and also colorants (labels). Sometimes amines are also added to lower the pH of the automotive fuel.

Other suitable fuel compositions of the invention comprise, in particular, blends of the gasoline described above with a mixture of gasoline additives containing the polar group (f) and corrosion inhibitors and/or lubricity improvers based on carboxylic acids or fatty acids, which can be present as monomeric and/or dimeric species. Typical mixtures of this type contain polyisobutylene amines combined with alkanol-initiated polyethers such as tridecanol or isotridecanol butoxylates or propoxylates, polyisobutylene amines combined with alkanol-initiated polyether amines such as reaction products of tridecanol or isotridecanol butoxylate with ammonia and alkanol-initiated polyether amines such as reaction products of tridecanol or isotridecanol butoxylate with ammonia combined with alkanol-initiated polyethers such as tridecanol or isotridecanol butoxylates or propoxylates, in each case combined with said corrosion inhibitors or lubricity improvers.

Said gasoline additives containing the polar groups (a) to (i) and said other components are metered to the gasoline, where they become effective. The components or additives can be added to the gasoline individually or as a previously prepared concentrate (additive pack).

Said gasoline additives containing the polar groups (a) to (i) are added to the gasoline usually in an amount of from 1 to 5000 ppm by weight, preferably from 5 to 3000 ppm by weight and more preferably from 10 to 1000 ppm by weight. The other compo-

nents and additives mentioned, if desired, are added in conventional amounts.

The fuel composition of the invention surprisingly allows for the
 5 use of distinctly less detergent or anti-valve-seat-wear agent to achieve the same detergent or pollution-abating action or anti-valve-seat-wear action as in the case of conventional fuel compositions of the prior art. Furthermore when the same amounts of detergent or anti-valve-seat-wear agent are used in the fuel com-
 10 position of the invention as in conventional fuel compositions there is achieved, surprisingly, a distinctly better detergent or pollution-abating or anti-valve-seat-wear action.

Furthermore, the fuel composition of the invention has additional
 15 advantages in that less sedimentation occurs in the combustion chamber of the Otto engine and less additive migrates to the motor oil due to fuel dilution.

The invention is illustrated by, but not restricted to, the fol-
 20 lowing examples.

Examples:

The gasolines used were those listed in Table 1 complying to the
 25 specifications stated, where OF 1 stands for a typical commercial Otto fuel.

Table 1

Grading	OF1 (for comparison)	OF2 (invention,)
aromatics content [vol%]	48.4	41.8
benzene content [vol%]	2.0	1.0
olefin content [vol%]	22.6	7.8
35 oxygen content [wt%]	0.5	1.7
sulfur content [ppm by weight]	245	90
summer vapor pressure (at 37°C) [kPa]	78.4	69.3

Preparation of the fuel compositions

Example 1 (comparative example)

- 5 700 mg of a polyisobutylene amine, prepared from highly reactive polyisobutylene having a molecular weight M_n of 1000 by hydroformylation and subsequent reductive amination with ammonia and dilution to equal parts by weight with C₁₀-C₁₄ paraffin (Kerocom® PIBA sold by BASF Aktiengesellschaft), were dissolved in 1 kg of
10 OF1 as indicated in Table 1.

Example 2 (invention)

- 700 mg of the same polyisobutylene amine as used in Example 1
15 were dissolved in 1 kg of OF2 as indicated in Table 1.

Example 3 (comparative example)

- 600 mg of a commercial additive formulation for gasolines, containing a conventional amount of a detergent containing carbamate groups as in group (f), were dissolved in 1 kg of OF1 as indicated in Table 1.

Example 4 (invention)

- 25 600 mg of the same commercial additive formulation for gasolines as used in Example 3 were dissolved in 1 kg of OF2 as indicated in Table 1.

30 Example 5 (comparative example)

- 400 mg of a commercial additive formulation for gasolines, containing a detergent, prepared by chlorination and subsequent amination of polyisobutylene having a molecular weight M_n of 950
35 and having predominantly centered double bonds, were dissolved in 1 kg of OF1 as indicated in Table 1.

Example 6 (invention)

- 40 400 mg of the same commercial additive formulation for gasolines as used in Example 5 were dissolved in 1 kg of OF2 as indicated in Table 1.

Example 7 (comparative example)

750 mg of a commercial additive formulation for gasolines, containing 50 wt% of the same polyisobutylene amine as used in Example 1 and also mineral and synthetic flotation oils and corrosion control agents (Keropur® 3222 sold by BASF Aktiengesellschaft) in conventional amounts, were dissolved in 1 kg of OF1 as indicated in Table 1.

10 Example 8 (invention)

350 mg of the same commercial additive formulation for gasolines as used in Example 7 were dissolved in 1 kg of OF2 as indicated in Table 1.

15 Example 9 (comparative example)

500 mg of a commercial additive formulation for gasolines, containing 60 wt% of the same polyisobutylene amine as used in Example 1 and also mineral flotation oil and corrosion control means (Keropur® 3233 sold by BASF Aktiengesellschaft) in conventional amounts, were dissolved in 1 kg of OF1 as indicated in Table 1.

Example 10 (invention)

25 500 mg of the same commercial additive formulation for gasolines as used in Example 9 were dissolved in 1 kg of OF2 as indicated in Table 1.

30 Example 11 (comparative example)

700 mg of a mixture of 50 wt% of the same polyisobutylene amine as used in Example 1 and 50 wt% of a commercial antiwear additive (Kerocom® 3280 sold by BASF Aktiengesellschaft) were dissolved in 35 1 kg of OF1 as indicated in Table 1.

Example 12 (invention)

700 mg of the same additive formulation for gasolines as used in 40 Example 11 were dissolved in 1 kg of OF2 as indicated in Table 1.

Working tests

Example 13 (comparative example)

- 5 Gasoline of Example 1 was examined as regards its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests employing a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no
10 additive is used, as shown in Table 2 below.

Example 14 (invention)

- Gasoline of Example 2 was examined as regards its suitability for
15 maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to find that
20 compared with Example 13 perfect cleaning of the inlet valves is achieved using the same amount of fuel additive.

Example 15 (comparative example)

- 25 Gasoline of Example 3 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no
30 additive is used, as shown in Table 2 below.

Example 16 (invention)

- Gasoline of Example 4 was examined to determine its suitability
35 for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to
40 find that compared with Example 15 virtually perfect cleaning of the inlet valves is achieved using the same amount of fuel additive.

Example 17 (comparative example)

Gasoline of Example 5 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below.

10 Example 18 (invention)

Gasoline of Example 6 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz 15 engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to find that compared with Example 17 virtually perfect cleaning of the inlet valves is achieved using the same amount of fuel additive. 20

Example 19 (comparative example)

25 Gasoline of Example 7 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below.

Example 30 (invention)

Gasoline of Example 8 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to find that distinctly less fuel additive is required than in Example 19 to achieve a similar degree of inlet valve cleanliness.

Example 21 (comparative example)

45 Gasoline of Example 9 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet

valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below.

Example 22 (invention)

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Gasoline of Example 10 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet

10 valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to find that compared with Example 21 distinctly better cleaning of the inlet valves is achieved using the same amount of fuel additive.

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Example 23 (comparative example)

Gasoline of Example 11 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying 20 out engine tests in the form of bench tests on a Mercedes-Benz engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below.

25 Example 24 (invention)

Gasoline of Example 12 was examined to determine its suitability for maintaining a clean inlet system. This was done by carrying out engine tests in the form of bench tests on a Mercedes-Benz 30 engine CEC F-05-A-93. As expected, the deposits on the inlet valves were distinctly less than the basic value obtained when no additive is used, as shown in Table 2 below. It was surprising to find that compared with Example 23 distinctly better cleaning of the inlet valves was achieved using the same amount of fuel additive.

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Table 2

Additive	Dosage [mg/kg]	Deposits on the inlet valves [mg/valve]					average
		valve 1	valve 2	valve 3	valve 4		
Ex. 13	700	40	157	7	87	73 (547)	
Ex. 14	700	0	0	0	0	0 (239)	
Ex. 15	600	19	60	86	34	50 (274)	
Ex. 16	600	0	1	0	2	1 (239)	
Ex. 17	400	0	75	17	182	69 (402)	
Ex. 18	400	0	2	2	0	1 (239)	
Ex. 19	750	31	120	111	30	73 (592)	
Ex. 20	350	46	68	38	67	55 (239)	
Ex. 21	500	181	95	26	68	93 (475)	
Ex. 22	500	27	33	14	77	38 (239)	
Ex. 23	700	123	12	98	55	72 (558)	
Ex. 24	700	82	12	23	22	35 (239)	

(the values in brackets refer to the basic value of the automotive fuel not containing any additive)

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We claim:

1. A fuel composition containing, as major component, a gasoline having an aromatics content of not more than 42 vol% and a sulfur content of not more than 150 ppm by weight, and, as minor component, at least one gasoline additive having a detergent action or an anti-valve-seat-wear action, wherein this gasoline additive contains at least one hydrophobic hydrocarbon group having a number-average molecular weight (M_n) of from 85 to 20,000 and at least one polar group selected from
 - 5 (a) monoamino or polyamino groups containing up to 6 nitrogen atoms, of which at least one has alkaline properties,
 - (b) nitro groups, optionally combined with hydroxyl groups,
 - (c) hydroxyl groups combined with monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties,
 - (d) carboxylic acid groups or the alkali metal or alkaline earth metal salts thereof,
 - (e) sulfo groups or the alkali metal or alkaline earth metal salts thereof,
 - (f) polyoxy-(C₂-C₄ alkylene) groups which are terminated by hydroxyl groups, monoamino or polyamino groups, in which at least one nitrogen atom has alkaline properties, or by carbamate groups,
 - (g) carboxylate groups,
 - (h) groups derived from succinic anhydride and containing hydroxyl and/or amino and/or amido and/or imido groups and
- 40 (i) groups produced by Mannich reaction of substituted phenols with aldehydes and mono- or poly-amines.
2. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (a), polyalkene mono-amine or polyalkene polyamines based on polypropylene, polybutylene or polyisobutylene having a molecular weight M_n of from 300 to 5000.

3. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (b), reaction products of polyisobutenes having an average degree of polymerization P of from 5 to 100 with nitrogen oxides or mixtures of nitrogen oxides and oxygen.
- 5
4. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (c), reaction products of polyisobutene epoxides, obtained from polyisobutylene containing predominantly terminal double bonds and having a molecular weight M_n of from 300 to 5000, with ammonia, mono- or poly-amines.
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5. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (d), copolymers of C₂-C₄₀olefins with maleic anhydride having a total molecular weight of from 500 to 20,000 whose carboxylic acid groups are completely or partially converted to the alkali metal or alkaline earth metal salts and the remainder of the carboxylic acid groups has been caused to react with an alcohol or amine.
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6. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (e), an alkali metal or alkaline earth metal salt of an alkyl sulfosuccinate.
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7. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (f), a polyether or polyether amine, obtainable by reaction of a C₂-C₃₀ alkanol, C₆-C₆₀ alkanediol, mono- or di-(C₂-C₃₀ alkyl)amine, C₁-C₃₀ alkylcyclohexanol or C₁-C₃₀ alkylphenol with from 1 to 30 mol of ethylene oxide and/or propylene oxide and/or butylene oxide per hydroxyl group or amino group and, in the case of polyether amines, by subsequent reductive amination with ammonia, a monoamine or a polyamine.
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8. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (g), an ester of a mono-, di- or tri-carboxylic acid with a long-chain alkanol or polyol.
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9. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (h), a derivative of polyisobutylenesuccinic anhydride, obtained by reaction of conventional or highly reactive polyisobutylene having a mo-
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lecular weight M_n of from 300 to 5000 with maleic anhydride by thermal treatment or via the chlorinated polyisobutylene.

10. A fuel composition as defined in claim 1, containing, as gasoline additive containing polar groups (i), a reaction product of a polyisobutene-substituted phenol with formaldehyde and a mono- or poly-amine.
- 10 11. A fuel composition as defined in any of claims 1 to 10, containing a gasoline having an olefin content of not more than 21 vol%.
- 15 12. A fuel composition as defined in any of claims 1 to 11, containing a gasoline having a benzene content of not more than 1.0 vol%.
13. A fuel composition as defined in any of claims 1 to 12, containing a gasoline having an oxygen content of not more than 2.7 wt%.
- 20 14. A fuel composition as defined in any of claims 1 to 13, containing the gasoline additives containing the polar groups (a) to (i) in a concentration of from 1 to 5000 ppm by weight.

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Fuel Composition

Abstract of the disclosure:

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A fuel composition containing, as major component, a gasoline having an aromatics content of not more than 42 vol% and a sulfur content of not more than 150 ppm by weight, and, as minor component, at least one gasoline additive having a detergent action or
10 an anti-valve-seat-wear action, in which this gasoline additive contains at least one hydrophobic hydrocarbon group having a number-average molecular weight of from 85 to 20,000 and at least one polar group.

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Declaration, Power of Attorney

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0050/049741

We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Fuel Composition

the specification of which

is attached hereto.

was filed on _____ as

Application Serial No. _____

and amended on _____.

was filed as PCT international application

Number PCT/EP00/00911

on February 5, 2000

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19905211.5	Germany	09 February 1999	[x] Yes [] No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

_____ (Application Number) _____ (Filing Date)

_____ (Application Number) _____ (Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

SCANNED # 24

Application Serial No.	Filing Date	Status (pending, patented, abandoned)

And we (I) hereby appoint **Messrs. HERBERT. B. KEIL**, Registration Number 18,967; and **RUSSEL E. WEINKAUF**, Registration Number 18,495; the address of both being Messrs. Keil & Weinkauf, 1101 Connecticut Ave., N.W., Washington, D.C. 20036 (telephone 202-659-0100), our attorneys, with full power of substitution and revocation, to prosecute this application, to make alterations and amendments therein, to sign the drawings, to receive the patent, and to transact all business in the Patent Office connected therewith.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Declaration

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0050/049741

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